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(71)出願人 000006013

三菱電機株式会社

東京都千代田区丸の内二丁目2番3号

(72)発明者 谷口 哲也

神戸市兵庫区和田崎町1丁目1番2号 三菱電機株式会社神戸製作所内

(72)発明者 松本 正昭

神戸市兵庫区和田崎町1丁目1番2号 三菱電機株式会社神戸製作所内

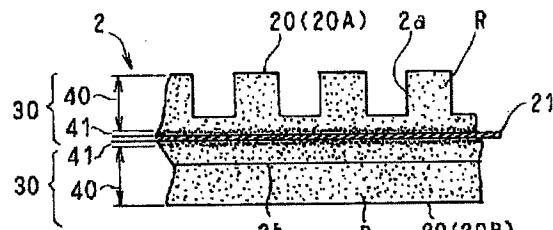
(74)代理人 弁理士 曾我 道照 (外6名)

(54)【発明の名称】 燃料電池用リブ付きセパレータおよびその製造方法

(57)【要約】

【目的】 電気抵抗や熱抵抗を小さく抑えることができるとともに、積層コストの低減を図ることができる燃料電池用リブ付きセパレータを提供する。

【構成】 その材料を、一定厚さの多孔質炭素層40の一側に薄くてシール性のある緻密質炭素層41を有した一体形の傾斜炭素板30、30の2枚から構成し、そのガス流路2a、2b用のリブRを、2枚の傾斜炭素板30、30の各多孔質炭素層40の他面側に形成する。この場合、リブRを形成した一对の傾斜炭素板30、30の緻密質炭素層41側どうしを重ね合わせるとリブ付きセパレータ2が形成されるが、接触面が一箇所しかないため、電気抵抗等を小さく抑えることができ、かつ積層コストの低減をも図ることができる。



30: 傾斜炭素材

40: 多孔質炭素層

41: 紹密質炭素層

# PATENT ABSTRACTS OF JAPAN

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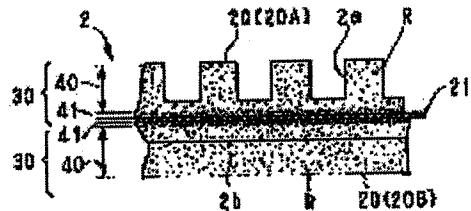
(72)Inventor : TANIGUCHI TETSUYA  
MATSUMOTO MASAAKI

## (54) SEPARATOR WITH RIB FOR FUEL CELL AND ITS MANUFACTURE

### (57)Abstract:

PURPOSE: To suppress electric resistance and thermal resistance small, and reduce layering cost.

CONSTITUTION: Material is composed of two integrated type inclined carbon plates 30, 30 having thin and sealing property dense carbon layers 41 on one sides of fixed thickness porous carbon layers 40. Ribs R for the gas paths 2a, 2b thereof are formed on the other sides of the respective porous carbon layers 40 of the two inclined carbon plates 30, 30. The respective dense carbon layer 41 sides of one pair of rib R-formed inclined carbon plates 30, 30 are superimposed so as to form a separator 2 with ribs R. Since there is only one touching face, it is possible to suppress electric resistance and the like small, and to reduce layering cost.



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## DETAILED DESCRIPTION

## [Detailed Description of the Invention]

## [0001]

[Industrial Application] This invention relates to the ribbed separator used for the phosphoric-acid form fuel cell of a laminated structure, and its manufacture approach.

## [0002]

[Description of the Prior Art] The phosphoric-acid form fuel cell of the laminating type which used the phosphoric acid as the electrolyte is known (JP,63-318075,A, JP,63-24561,A, JP,60-230366,A, JP,60-20471,A, JP,57-19981,A, etc.). For example, drawing 13 is the decomposition perspective view of the layered product of such a conventional laminating type phosphoric-acid form fuel cell, and is set to drawing. The cell which found both sides of matrix 1a into which the phosphoric acid with which 1 becomes an electrolyte was infiltrated by air electrode 1b as an electrode, and fuel electrode 1c, It is the layered product to which 2 has level transverse groove-like airstream way 2a as a gas passageway in a whole surface side, and the ribbed separator of both quirks with level fluting-like fuel gas passage 2b and 3, on the other hand, carried out two or more laminatings of a cell 1 and the ribbed separator 2 to the side by turns as a gas passageway.

[0003] Drawing 14 is the expanded sectional view of the above-mentioned ribbed separator 2, and is set to drawing. The porous carbon plate with which 10 has gas permeability, and 11 have vapor-liquid seal nature. The substantia-compacta carbon plate inserted with the porous carbon plates 10 and 10 of a pair, The rib with which R is formed in the external surface side of the porous carbon plate 10 as an object for fuel gas passage 2b as an object for airstream way 2a, and R1 are the seal ribs with which the seal processing for preventing leak of the vapor-liquid from the both-sides side of the porous carbon plate 10 was made. Since the board thickness of the substantia-compacta carbon plate 11 is usually 0.4-1.0mm here and the board thickness of the porous carbon plate 10 is usually 1.0-2.0mm, the thickness of the ribbed separator 2 whole is set to 2.4-5.0mm. Moreover, since the height of Rib R is usually about 0.5-1.5mm, the reserved meat thickness for a slot of the porous carbon plate 10 is set to about 0.5mm.

[0004] Actuation of this laminating type phosphoric-acid form fuel cell is explained below. air supplies airstream way 2a of a ribbed separator 2 through the gas header (not shown) prepared in the side face of a layered product 3 in the generation of electrical energy -- having -- fuel gas passage 2b -- hydrogen -- rich fuel gas is supplied. And this air and fuel gas reach air electrode 1b of a cell 1, and fuel electrode 1c, respectively, diffusing the inside of the porous carbon plate 10 of direct or a ribbed separator 2, and cause a reaction under each catalyst. While the hydrogen ion generated in fuel electrode 1c at that time moves in the inside of matrix 1a and reaching to air electrode 1b, the electron generated in fuel electrode 1c reaches to air electrode 1b of the next cell 1 through the conductive ribbed separator 2, and water is made to generate by this air electrode 1b. And electromotive force occurs for every cell at this time, and only the electromotive force which multiplied the electromotive force of a cell 1 by the number of cells 1 occurs in the layered product 3 whole.

[0005] In this case, although it has work that a through electron does not let only a hydrogen ion pass, according to an operation of the phosphoric acid whose matrix 1a is an electrolyte, the phosphoric acid

in this matrix 1a is taken out out of a fuel cell subsystem in part at the time of evaporation of the generated water etc., and serves as insufficient feeling. For this reason, if the phosphoric acid is beforehand stored all over the opening of the porous carbon plate 10 of a ribbed separator 2 and phosphoric acids run short by the matrix 1a side, this phosphoric acid is continuously supplied from the porous carbon plate 10 side of a ribbed separator 2. Moreover, although heat occurs in cell 1 grade by the electromotive loss at the time of a generation of electrical energy and it is going to raise the temperature in a fuel cell subsystem, this heat passes along a ribbed separator 2, is transmitted to the cooling system (not shown) arranged for every number cell, and is collected from there outside.

[0006] While a laminating type phosphoric-acid form fuel cell has conductivity and high thermal conductivity nature to a ribbed separator 2 in order to operate as mentioned above, it is required that it should have thermal resistance and a phosphoric-acid-proof corrosive. And it is required for the substantia-compacta carbon plate 11 of a ribbed separator 2 with high vapor-liquid seal nature and high intensity that thinning should be possible, and it is required especially that gas permeability and phosphoric-acid keeping should be good for the porous carbon plate 10 of a ribbed separator 2. For this reason, the amorphous carbon ingredient (for example, SG series by Showa Denko K.K. and the GCR series by Kobe Steel, Ltd. correspond to this) called common-name glassy carbon is conventionally used for the substantia-compacta carbon plate 11 by the thickness which is about 0.4-0.8mm. To the porous carbon plate 10, a pitch system or a PAN system, Or the carbon ingredient (for example, the KES series by Kureha Chemical Industry Co., Ltd. and the TGP series by Toray Industries, Inc. correspond to this) calcinated after mixing and fabricating other fiber and thermosetting resin is used by the thickness which is 1-2mm.

[0007] Now, when the operation of the ribbed separator 2 which consists of a porous carbon plate 10 and a substantia-compacta carbon plate 11 is classified, the porous carbon plate 10 and the substantia-compacta carbon plate 11 are used as an independent member, and there is the approach of carrying out the laminating of what joined or unified beforehand the approach of carrying out the laminating separately at the time of assembly, and two porous carbon plates 10 and 10 and one substantia-compacta carbon plate 11.

[0008] To the approach of joining or unifying, the porous carbon plate 10 and the substantia-compacta carbon plate 11 here A : The junction approach which applies cement to what baking ended, respectively, pressurizes and heat-treats this etc. after that, and is stuck, B: The unification approach of pressurizing and calcinating this etc. after applying thermosetting resin to what baking ended, respectively and sticking it on it, and after C:one side or both sides pressurizes the things non-calcinated (the so-called Green condition is said) and sticks, there is the unification approach of calcinating this etc.

However, since a difference is in whenever [ coefficient-of-linear-expansion, elastic-coefficient, and contraction ] between the porous carbon plate 10 and the substantia-compacta carbon plate 11 in fact, the unification approach of A and B is difficult, and many junction approaches of C are adopted.

[0009] By the approach of Above C, the admixture of a fluororesin system or this, and a carbon material etc. is used from a viewpoint of the ingredient which has phosphoric-acid-proof nature as cement. And after applying this cement to the front face of the substantia-compacta carbon plate 11 thinly and drying, on both sides of this substantia-compacta carbon plate 11, it pressurizes with two porous carbon plates 10 and 10, the temperature up of this etc. is carried out to 300-400 degrees C, and the ribbed separator 2 as a zygote is manufactured. In addition, the ribbed separator of a piece quirk should just join similarly one porous carbon plate 10 and one substantia-compacta carbon plate 11.

[0010]  
[Problem(s) to be Solved by the Invention] However, when it constitutes a ribbed separator 2 from two porous carbon plates 10 and one substantia-compacta carbon plate 11 which became independent separately, there is a merit that it is not necessary to join this etc. beforehand and manufacture cost is cheap, but since the count of a part laminating increases, the technical problem that the rise of laminating cost will be caused occurs. Moreover, in order to have to carry out the laminating of the low porous carbon plate 10 of plate rigidity which carried out recessing in this case independently, the

technical problem that it is easy to produce breakage is in this porous carbon plate 10 at the time of handling. For this reason, a skilled craftsman's handicraft must perform the laminating of the porous carbon plate 10, and the technical problem that the rise of that part assembly cost will be caused also occurs. In addition, although heavy-gage-izing and densification can be considered to aim at improvement in plate rigidity of the porous carbon plate 10, this is not contrary and appropriate to the conductive and thermally conductive improvement required of the porous carbon plate 10, or the improvement in gas permeability while it is contrary to miniaturization of a cell.

[0011] Moreover, in carrying out the laminating of the porous carbon plate 10 or the substantia-compacta carbon plate 11 separately, corresponding to the planar pressure relaxation by planar pressure distribution and creep deformation of a layered product 3, it is easy to produce big electric resistance and thermal resistance into a contact part locally, and the technical problem that a battery life will be shortened occurs.

[0012] On the other hand, when two porous carbon plates 10 and one substantia-compacta carbon plate 11 are joined in one and it constitutes a ribbed separator 2 Although there is naturally a demerit of it being necessary to the above-mentioned case to do the junction activity of this etc. on the merit that the count of a laminating decreases sharply in the assembly of a layered product 3, beforehand, and causing the rise of manufacture cost In order to use fluororesin as cement especially in this case, in the part (part to which the porous carbon plate 10 and the substantia-compacta carbon plate 11 are not completely close) whose junction condition is not good, the technical problem that that conductivity and thermal conductivity will fall occurs. Moreover, even if electric resistance and thermal resistance have become below the allowed value as the one whole zygote, since it is large, dispersion in a plane of composition has in fact the technical problem that this dispersion will also affect the way and temperature distribution to which the current within a cell 1 flows, and will finally have a bad influence on the property of a cell 1, or the generating efficiency of a cell.

[0013] Furthermore, although it is necessary in junction to heat to the temperature which can fuse fluororesin, since the temperature distribution within the plate surface at the time of heating affect bonding strength as it is, there is an inclination heated from need temperature to high temperature. For this reason, a part of fluororesin evaporates, it is spread in the porous carbon plate 10, a carbon fiber front face is covered, and the technical problem that the phosphoric-acid keeping of the porous carbon plate 10 will worsen occurs.

[0014] Moreover, when joining a ribbed separator 2 and really considering as a form, also by the case, with this ribbed separator 2, contact or a junction part is generated about both sides of the substantia-compacta carbon plate 11, and the technical problem that electric resistance and thermal resistance will be greatly increased in this part occurs. And in the layered product 3 whole, since contact or the junction part of this ribbed separator 2 serves as an immense number, it has the technical problem that this etc. has a bad influence on the generating efficiency of a fuel cell, or cooling of a layered product 3.

[0015] It aims at offering the ribbed separator for fuel cells which can aim at reduction of laminating cost while this invention can be made, and can fulfill the engine performance and quality demanded and can stop electric resistance and thermal resistance small, in order to solve the above technical problems. Moreover, this invention aims at offering the manufacture approach of the ribbed separator for fuel cells that the ribbed separator for the above-mentioned fuel cells can be manufactured with the sufficient yield by low cost.

[0016]

[Means for Solving the Problem] In the ribbed separator for fuel cells with which the laminating of the invention of the 1st of this invention is carried out to the both sides holding an electrolyte of a matrix by turns [ a cell and by turns ] with an electrode, and the rib for gas passageways is formed in those both sides The ingredient is really with the substantia-compacta carbon layer which is thin and has seal nature in the whole surface side of the porous carbon layer of fixed thickness constituted from two formal inclination carbon plates, and it is the thing of each porous carbon layer of two porous carbon plates currently formed in a side on the other hand about the rib for the gas passageways.

[0017] In the manufacture approach of the ribbed separator for fuel cells that the laminating of the

invention of the 2nd of this invention is carried out to the both sides holding an electrolyte of a matrix by turns [ a cell and by turns ] with an electrode, and the rib for gas passageways is formed in those both sides The 1st process which manufactures the porous carbon plate of the fixed thickness which has a rib for gas passageways in a whole surface side, It is having the 2nd process of a porous carbon plate which, on the other hand, forms the thin film of thermosetting resin in a side, and the 3rd process which heat-treats the porous carbon plate with which the thin film's was formed, and changes a thin film into a substantia-compacta carbon layer with seal nature.

[0018] In the ribbed separator for fuel cells with which the laminating of the invention of the 3rd of this invention is carried out to the both sides holding an electrolyte of a matrix by turns [ a cell and by turns ] with an electrode, and the rib for gas passageways is formed in those both sides It is really with the substantia-compacta carbon layer which is thin and has seal nature between the porous carbon layers of the pair of fixed thickness constituting the ingredient from a formal inclination carbon plate, and forming the rib for the gas passageways in both the external surface side of the porous carbon layer of an inclination carbon plate.

[0019] In the manufacture approach of the ribbed separator for fuel cells that the laminating of the invention of the 4th of this invention is carried out to the both sides holding an electrolyte of a matrix by turns [ a cell and by turns ] with an electrode, and the rib for gas passageways is formed in those both sides The 1st process which manufactures the porous carbon plate of the fixed thickness which has a rib for gas passageways in a whole surface side, It is having the 2nd process of a porous carbon plate which, on the other hand, forms the thin film of thermosetting resin in a side, and the 3rd process which changes the thin film sides of the porous carbon plate of a pair with which the thin film's was formed into the superposition and substantia-compacta carbon layer which has seal nature in a thin film while heat-treating and unifying this etc.

[0020] In the ribbed separator for fuel cells with which the laminating of the invention of the 5th of this invention is carried out to the both sides holding an electrolyte of a matrix by turns [ a cell and by turns ] with an electrode, and the rib for gas passageways is formed in those both sides The ingredient is really with the substantia-compacta carbon layer which is thin and has seal nature in the whole surface side of the porous carbon layer of fixed thickness constituted from two formal inclination carbon plates. It is forming the rib for the quantities of gas flow in both the external surface side of the porous carbon layer of two inclination carbon plates which joined substantia-compacta carbon layer sides and were unified.

[0021] In the manufacture approach of the ribbed separator for fuel cells that the laminating of the invention of the 6th of this invention is carried out to the both sides holding an electrolyte of a matrix by turns [ a cell and by turns ] with an electrode, and the rib for gas passageways is formed in those both sides The 1st process which manufactures the porous carbon plate of the fixed thickness which has a rib for gas passageways in a whole surface side, The 2nd process of a porous carbon plate which, on the other hand, forms the thin film of thermosetting resin in a side, It is having the 3rd process which heat-treats the porous carbon plate with which the thin film's was formed, and changes a thin film into a substantia-compacta carbon layer with seal nature, and the 4th process which joins the substantia-compacta carbon layer sides of the porous carbon plate of a pair with which the substantia-compacta carbon layer's was formed, and is unified.

[0022] In the ribbed separator for fuel cells with which the laminating of the invention of the 7th of this invention is carried out to the both sides holding an electrolyte of a matrix by turns [ a cell and by turns ] with an electrode, and the rib for gas passageways is formed in those both sides The ingredient really with the substantia-compacta carbon layer which is thin and has seal nature in the whole surface side of the porous carbon layer of fixed thickness Two formal inclination carbon plates, It is constituting from a carbon plate with thin reinforcement, and forming the rib for quantities of gas flow in porous carbon \*\*\*\*\* external surface, although the carbon plate's was inserted by the substantia-compacta carbon layer sides of two inclination carbon plates and coalesce junction was carried out.

[0023] In the manufacture approach of the ribbed separator for fuel cells that the laminating of the invention of the 8th of this invention is carried out to the both sides holding an electrolyte of a matrix by turns [ a cell and by turns ] with an electrode, and the rib for gas passageways is formed in those both

sides The 1st process which manufactures the porous carbon plate of the fixed thickness which has a rib for gas passageways in a whole surface side, The 2nd process of a porous carbon plate which, on the other hand, forms the thin film of thermosetting resin in a side, The 3rd process which heat-treats the porous carbon plate with which the thin film was formed, and changes a thin film into a substantia-compacta carbon layer with seal nature, It is having the 4th process which sandwiches a carbon plate with thin reinforcement by the substantia-compacta carbon layer sides of the porous carbon plate of a pair with which the substantia-compacta carbon layer's was formed, joins, and unifies this etc.

[0024]

[Function] In invention of the 1st of this invention, a ribbed separator is obtained by piling up two inclination carbon plates (henceforth a half separator) with which the rib for gas passageways was formed in the porous carbon layer side. That is, since this half separator really which has the substantia-compacta carbon layer which is thin and has seal nature in the 1 side of a porous carbon layer consists of formal inclination carbon plates, a ribbed separator is obtained by piling up the mutual two substantia-compacta carbon layer side of a half separator. Moreover, since a ribbed separator is formed with the half separator of two sheets, only one place becomes easy [ the laminating activity ], even when it forms a layered product with a cell, while there is no contact part and it can stop electric resistance and thermal resistance small. Furthermore, since the half separator 20 has the substantia-compacta carbon layer, improvement in simple substance reinforcement can be aimed at, and reduction of the breakage at the time of handling is achieved.

[0025] the porous carbon plate of fixed thickness with the rib for [ at the 1st process / to a whole surface side ] gas passageways by invention of the 2nd of this invention -- manufacturing -- the 2nd process -- this porous carbon plate -- on the other hand, the thin film of thermosetting resin is formed in a side. And the porous carbon plate with which the thin film was formed at the 3rd process is heat-treated, and this thin film is changed into a substantia-compacta carbon layer with seal nature. That is, the half separator concerning the 1st invention is manufactured according to processes, such as this. At processes, such as this, a thin substantia-compacta carbon layer cannot be divided and can be formed in the 1 side of a porous carbon plate.

[0026] In invention of the 3rd of this invention, the ribbed separator is formed by forming the rib for gas passageways in the external surface side of the porous carbon layer which it has on both sides of an inclination carbon plate. Since the inclination carbon plate in this case has in one the substantia-compacta carbon layer which is thin and has seal nature between the porous carbon layers of the pair of fixed thickness, if the rib for gas passageways is formed in those both sides, it will serve as a ribbed separator. It becomes still easier [ the laminating activity in the case of forming a layered product ] while a contact part cannot be generated but it can lower electric resistance and thermal resistance from the ribbed separator of the 1st invention at the time of a laminating, since this ribbed separator is really a formal thing. Moreover, the breakage at the time of handling can also be reduced further.

[0027] In invention of the 4th of this invention, although the 1st process and 2nd process are the same as that of it of the 2nd invention, it is the 3rd process and said thin film sides of the porous carbon plate of a pair with which the thin film was formed are changed into superposition and the substantia-compacta carbon layer which has seal nature in said thin film while heat-treating and unifying these. That is, the ribbed separator 2 concerning the 3rd invention is manufactured according to these processes. At these processes, between the porous carbon plates of a pair, a thin substantia-compacta carbon layer cannot be divided and can be formed.

[0028] It is the case where joined the substantia-compacta carbon layer side of the half separator of the pair which starts the 1st invention in a ribbed separator, and it constitutes from invention of the 5th of this invention in one, and from the case of the ribbed separator of the 1st invention, the laminating activity in the case of forming a layered product becomes easy, and the breakage at the time of handling is also reduced further.

[0029] In invention of the 6th of this invention, the 1st process to the 3rd process is the same as that of it of the 2nd invention, at the 4th process, the substantia-compacta carbon layer side of the half separator of a pair is joined, and the ribbed separator of a junction type is obtained. That is, the ribbed separator

which starts the 5th invention according to these processes is manufactured.

[0030] By inserting the carbon plate which is thin and has reinforcement in invention of the 7th of this invention between substantia-compacta carbon layer the half separator of the pair concerning the 1st invention sides, joining, and making three persons coalesce in one, it is the case where a ribbed separator is formed, and from the case of the ribbed separator of the 5th invention, that reinforcement is raised and further prevention of the breakage at the time of handling is achieved.

[0031] In invention of the 8th of this invention, the 1st process to the 3rd process is the same as that of it of the 2nd invention, and at the 4th process, the carbon plate which is thin and has reinforcement is inserted, it joins to the substantia-compacta carbon layer side of the half separator of a pair, and the ribbed separator of a junction type is obtained. That is, the ribbed separator which starts the 7th invention according to these processes is manufactured.

[0032]

[Example] Hereafter, the example of this invention is explained about drawing.

example 1. -- one example of the ribbed separator for fuel cells concerning the 1st invention of this invention is explained with reference to drawing 1 thru/or drawing 3 . Drawing 1 is [ the partial sectional view of a ribbed separator and drawing 3 of the partial perspective view of the layered product of a laminating type phosphoric-acid form fuel cell and drawing 2 ] the sectional views of an inclination carbon plate. In addition, the same sign is given to the same as that of the layered product of the conventional laminating type phosphoric-acid form fuel cell shown by drawing 13 , or a considerable part, and the explanation is omitted.

[0033] In drawing, 20 is the half separator (piece quirk ribbed separator) with which Rib R was formed in the whole surface side, and the half separator for air electrodes with which, as for 20A, the rib R for airstream way 2a was formed by the whole surface side, and 20B are the half separators for fuel gas electrodes with which the rib R for fuel gas passage 2b was formed in the sense which intersects perpendicularly with the rib R for airstream way 2a at a whole surface side. 21 is a conductive graphite paste applied between them, when piling up half separator 20A for air electrodes, and half separator 20B for fuel gas electrodes and forming a ribbed separator 2.

[0034] As the half separator 20 is shown by drawing 3 , the ingredient consists of inclination carbon plates 30 of an one form with a thickness of 1.5mm with the substantia-compacta carbon layer 41 which has 100-200-micrometer thin vapor-liquid seal nature at the whole surface side of the porous carbon layer 40 with the permeability of predetermined thickness, and the rib R is formed in the field by the side of the porous carbon layer 40 in height of 1mm. since [ in addition, ] that reserved meat thickness is set to 0.5mm in this case -- the trough of Rib R -- up to the substantia-compacta carbon layer 41 -- pile \*\*\*\* -- there are nothings. And the ribbed separator 2 with a thickness of 3mm is formed by piling up the substantia-compacta carbon layer 41 side faces of two half separators 20A and 20B through the graphite paste 21. In addition, the rib R of the both sides of the half separator 20 is the seal rib R1 with which gas-seal processing was made.

[0035] Actuation of ribbed separator 2A which doubles two half separators 20A and 20B which consist of this inclination carbon plate 30 next, and is formed is explained. Since the half separator 20 consists of inclination carbon plates 30 which consist of a porous carbon layer 40 and a substantia-compacta carbon layer 41, it has all the engine performance and quality of the porous carbon plate 10 which constitutes the conventional ribbed separator 2, and the substantia-compacta carbon plate 11 on the property. Therefore, the ribbed separator 2 which piles up this half separator 20A for air electrodes and half separator 20B for fuel gas electrodes through the graphite paste 21, and is formed has the same function as the conventional ribbed separator 2, and the actuation of it at the time of that generation of electrical energy is the same as that of the conventional ribbed separator 2.

[0036] \*\*\*\*\* and the conventional ribbed separator 2 consist of three components of two porous carbon plates 10 and 10 and one substantia-compacta carbon plate 11. Since the ribbed separator 2 of this example consists of two half separators 20A and 20B to that contact or a junction part being two places and there is that contact part of a, The electric resistance and thermal resistance were reduced remarkably, and it was confirmed by the experiment that these values have become or less about 1 of the

value of the conventional ribbed separator 2 / 2. Moreover, the various troubles which originate in a contact part with reduction of a contact part can be decreased.

[0037] Moreover, the conventional ribbed separator 2 with which two porous carbon plates 10 and 10 and one substantia-compacta carbon plate 11 are separately constituted independently is received. Since the ribbed separator 2 of this example 1 consists of two half separators 20A and 20B, while being able to aim at reduction of that laminating cost in the assembly of a layered product 3 Since the flexural strength of the half separator 20 is the part which has the substantia-compacta carbon layer 41, and twice [ more than ] the conventional porous carbon plate 10, breakage of the components at the time of handling can also be decreased sharply. Therefore, the assembly by persons other than a skilled craftsman also becomes possible. Furthermore, in the ribbed separator 2 of this example, it compares with the above-mentioned conventional ribbed separator 2, and is hard coming to generate big electric resistance and thermal resistance into a contact part locally corresponding to the planar pressure relaxation by the planar pressure distribution and creep deformation of a part and a layered product 3 whose contact part at the time of a laminating decreases, and a battery life can be lengthened so much.

[0038] Moreover, two porous carbon plates 10 and 10 and one substantia-compacta carbon plate 11 are joined, to the conventional ribbed separator 2 constituted by coalescing, in the ribbed separator 2 of this example, since there is no junction part, the part whose junction condition is not good does not arise, and big dispersion arises neither in electric resistance nor thermal resistance within a ribbed separator 2. Furthermore, in the ribbed separator 2 of this example 1, since there is no adhesion part, cement (fluororesin) is spread in the porous carbon layer 40, and does not worsen phosphoric-acid keeping of this porous carbon layer 40.

[0039] Example 2. drawing 4 is drawing showing one example of the manufacture approach of the ribbed separator for fuel cells concerning invention of the 2nd of this invention. Although two half separators 20A and 20B are piled up and the ribbed separator 2 is constituted from an example 1, this example 2 is related with the manufacture approach of this half separator 20.

[0040] This manufacture approach heat-treats the porous carbon plate with which the 1st process A1 which manufactures the porous carbon plate of the fixed thickness which has the rib R for gas passageways in a whole surface side, the 2nd process A2 of said porous carbon plate which, on the other hand, forms the thin film of thermosetting resin in a side, and said thin film were formed, and consists of the 3rd process A3 which changes said thin film into the substantia-compacta carbon layer 41 with seal nature.

[0041] The 1st process A1 is a process which manufactures the porous carbon plate which has the rib R for gas passageways in the whole surface side known from the former (for example, the approach is explained by JP,59-68170,A in full detail). After mixing a short carbon fiber, a phenol resin binder, and an organic particulate matter at this process, for example, After heating and pressurizing this, considering as the mold goods of fixed thickness and calcinating these mold goods at 2000 degrees C after that, The porous carbon plate which has a rib for gas passageways in a whole surface side is manufactured by processing the made porous carbon material (this serving as KES-400 and the equivalent device for example, by Kureha Chemical Industry Co., Ltd.) into predetermined size, and carrying out rib processing further. In addition, configuration processing containing Rib R may be performed in the phase of mold goods.

[0042] After the 2nd process A2 develops \*\*\*\* of thermosetting resin (for example, the poly carbodiimide resin by NISSHINBO INDUSTRIES, INC.) on a smooth glass plate, removes a solvent and obtains the thin film of thermosetting resin, it develops to the field of the side which does not have the rib R of the porous carbon plate which was able to do this thin film at said 1st process A1, and it dries this. 3rd process A3 carries out the temperature up of the porous carbon plate of the 2nd process A2 with which the thin film was developed and dried to the whole surface side gradually from a room temperature to 600 degrees C in a nitrogen ambient atmosphere, and cools it radiationally immediately after that. A thin film is calcinated by this, and it becomes a substantia-compacta carbon layer and adheres to the whole surface side of a porous carbon plate in one. And seal processing of the rib R of both ends is carried out, and the seal rib R1, then the half separator 20 the board thickness of whose is

1.5mm are completed.

[0043] In this case, the thickness of the substantia-compacta carbon layer formed from this thin film is 100-200 micrometers. Since it is thin, while there is also little reduction of the opening volume of the porosity part of a porous carbon plate That value of the gas-seal engine performance of this substantia-compacta carbon layer is below  $1.0 \times 10^{-6}$  (N-cc/min/cm<sup>2</sup>) [N<sub>2</sub> gas and differential pressure 0.2 kg/cm<sup>2</sup> and a room temperature], and since it is very high, a problem is not produced for the vapor-liquid seal engine performance of the direction of board thickness of this substantia-compacta carbon layer. In addition, the approach of forming thin substantia-compacta carbon material from poly carbodiimide resin is explained by JP,2-152167,A in full detail.

[0044] While the inclination carbon plate 30 from which the porous carbon plate became the porous carbon layer 40, and the thin film of thermosetting resin became the substantia-compacta carbon layer 41 by the above process [ 1st, 2nd, and 3rd ] A1 and A2 and A3 is manufactured, the same half separator 20 as the half separator 20 of an example 1 with which the rib R for gas passageways was formed in the porous carbon layer 40 side of this inclination carbon plate 30 is manufactured with the sufficient yield by low cost. In addition, half separator 20A for air electrodes and half separator 20B for fuel gas electrodes are separately manufactured according to the above-mentioned process in this case.

[0045] here -- substantia-compacta carbon material -- manufacturing -- hitting -- especially, contraction at the time of baking -- large -- and the compactness -- securing -- board thickness -- fixed thickness (0.4-1.0mm) -- until -- since it had to be made heavy-gage, it was difficult by generating of a crack to form a substantia-compacta carbon layer in the 1 side of porous carbon material [ finishing / baking ] (1.5mm) by approach like 3rd process A3. It is thought that it can form a substantia-compacta carbon layer small [ contraction at the time of the baking ], without producing a crack by 3rd process A3 since the burning temperature is comparatively as low as 600 degrees C while being able to fabricate

\*\*\*\*\*\*, for example, poly carbodiimide resin, to light-gage (200 micrometers following) very much.

[0046] In addition, although the poly carbodiimide resin by NISSHINBO INDUSTRIES, INC. is used as thermosetting resin in this example 2, if not only this but a thin substantia-compacta carbon layer can be broken and formed in the 1 side of a porous carbon plate [ finishing / baking ], it is needless to say [ thermosetting resin ] that other things may be used.

[0047] example 3. -- one example of the ribbed separator for the 3rd invention \*\*\*\* fuel cells of this invention is explained with reference to drawing 5 thru/or drawing 7 . Drawing 5 is [ the partial sectional view of a ribbed separator and drawing 7 of the partial perspective view of the layered product of a laminating type phosphoric-acid form fuel cell and drawing 6 ] the sectional views of an inclination carbon plate. In addition, the same sign is given to the same as that of the layered product of the laminating type phosphoric-acid form fuel cell of an example 1, or a considerable part, and the explanation is omitted.

[0048] The ribbed separator 2 of this example 3 is 200-400 micrometers between two porous carbon layers 40 with [ as it was really a form thing and that ingredient was shown by drawing 7 ] the permeability of predetermined thickness, and 40. It consists of inclination carbon plates 31 of an one form with a thickness of 3.0mm with the substantia-compacta carbon layer 42 which has thin vapor-liquid seal nature. And two or more ribs R with a height of 1mm are formed in both the external surface of this inclination carbon plate 31, i.e., each external surface of the porous carbon layers 40 and 40, and while fuel gas passage 2b is formed in one field of this inclination carbon plate 31 in the field of airstream way 2a and another side, the rib R of both ends is the seal rib R1 with which gas-seal processing was made.

[0049] Therefore, while really having the engine performance and the quality as the conventional ribbed separator 2 which consists of this inclination carbon plate 31 also with the same formal ribbed separator 2, it has the same function. Moreover, since there is no contact part in that interior since this ribbed separator 2 consists of one simple substance, and there is no increase of the electric resistance in this part or thermal resistance, that electric resistance and thermal resistance have become 1/10 or less [ of the conventional ribbed separator 2 ]. Furthermore, since the ribbed separator 2 of an example 1 does not have a contact part, other various troubles resulting from this contact part do not produce it.

[0050] Moreover, it is unnecessary, and can also aim at reduction of that part manufacture cost while it can aim at reduction of that laminating cost compared with the ribbed separator 2 of an example 1, since the ribbed separator 2 of this example 3 consists of one simple substance. [ of the graphite paste 21 ] Furthermore, that the vapor-liquid seal nature of the ribbed separator 2 of an example 3, flexural strength, and the opening volume of a porosity part are equivalent to the ribbed separator 2 of an example 1 or having the engine performance beyond it are checked in the evaluation test.

[0051] Example 4. drawing 8 is drawing showing one example of the manufacture approach of the ribbed separator for fuel cells concerning invention of the 4th of this invention. This example 4 is related with the manufacture approach of the ribbed separator 2 of the one form of an example 3.

[0052] The 1st process B1 at which this manufacture approach manufactures the porous carbon plate of the fixed thickness which has the rib R for gas passageways in a whole surface side, While piling up and heat-treating said thin film sides of the porous carbon plate of a pair with which said thin film was formed and uniting these with 2nd process B-2 of said porous carbon plate which, on the other hand, forms the thin film of thermosetting resin in a side It consists of the 3rd process B3 which changes said thin film into a substantia-compacta carbon layer with seal nature.

[0053] Since the 1st process B1 and 2nd process B-2 are the same as that of the 1st process A1 of an example 2, and the 2nd process A2 respectively, the explanation is omitted. At the 3rd process B3, as the thin films are set, immediately after piling up the porous carbon plate formed in the whole surface side by 2nd process B-2 of the pair by which the thin film was developed and dried, in a nitrogen ambient atmosphere, from a room temperature to 600 degrees C, the temperature up of these is carried out gradually, and they are cooled radiationally after that. While a thin film is calcinated by this, becoming a substantia-compacta carbon layer at the whole surface side of a porous carbon plate and adhering in one, substantia-compacta carbon layers coalesce. And seal processing of the predetermined rib R of this unified carbon plate is carried out, and the seal rib R1, then the ribbed separator 2 that board thickness of whose is 3.0mm are completed.

[0054] While the inclination carbon plate 31 from which two porous carbon plates became the porous carbon layers 40 and 40 of a pair, and the thin film of two thermosetting resin became the substantia-compacta carbon layer 42 by the above processes [ 1st, 2nd, and 3rd ] B1, B-2, and B3 is manufactured. The same ribbed separator 2 as the ribbed separator 2 of an example 3 with which the rib R for gas passageways was formed in the external surface side of the porous carbon layers 40 and 40 of the pair of this inclination carbon plate 31 is manufactured with the sufficient yield by low cost. In addition, in this case, the sense of the rib R by the side of one field of the inclination carbon plate 31 and the sense of the rib R by the side of the field of another side lie at right angles, airstream way 2a is formed in one field side of the inclination carbon plate 31, and fuel gas passage 2b is formed in the field side of another side.

[0055] Example 5. drawing 9 is drawing showing one example of the ribbed separator for fuel cells concerning invention of the 5th of this invention. The ribbed separator 2 of this example 5 joins the mutual substantia-compacta carbon layer 41 sides of half separator 20A for air electrodes of an example 1, and half separator 20B for fuel gas electrodes through the heat-resistant adhesives 22 of entering [ which has conductivity ] a graphite (for example, thermostat cement by the British salmon company etc.), and is unified. in addition, the thickness of this ribbed separator 2 -- the thickness of the half separators 20A and 20B -- respectively -- 1.5mm -- it is -- the thickness of the heat-resistant adhesives 22 -- 100 micrometers it is -- a sake -- 3.1mm -- becoming .

[0056] It can aim at improvement in reinforcement while it can aim at reduction of laminating cost from the ribbed separator 2 of a part to have joined and unified while being able to acquire the same effectiveness as the ribbed separator 2 of an example 1, and an example 1, since the ribbed separator 2 of this example 5 joins the half separators 20A and 20B of the pair of an example 1 and unifies.

[0057] In addition, the thing is [ that what is necessary is just what does not spoil not only this but a conductive function, but has thermal resistance ] natural although the thermostat cement by the British salmon company is used as heat-resistant adhesives 22 in this example 5.

[0058] Example 6. drawing 10 is drawing showing one example of the manufacture approach of the

ribbed separator for fuel cells concerning invention of the 6th of this invention. This example 6 is related with the manufacture approach of the ribbed separator of the junction type of an example 5.

[0059] The 1st process C1 at which this manufacture approach manufactures the porous carbon plate of the fixed thickness which has the rib R for gas passageways in a whole surface side, The 2nd process C2 of said porous carbon plate which, on the other hand, forms the thin film of thermosetting resin in a side, The 3rd process C3 which heat-treats the porous carbon plate with which said thin film was formed, and changes said thin film into a substantia-compacta carbon layer with seal nature, It consists of the 4th process C4 which joins said substantia-compacta carbon layer sides of the porous carbon plate of a pair with which said substantia-compacta carbon layer 41 was formed, and is unified.

[0060] Since the 1st process C1 to the 3rd process C3 is the same as that of the 1st process A1 of an example 2 to 3rd process A3 respectively, the explanation is omitted. After an organic solvent washes the field by the side of [ mutual ] the substantia-compacta carbon layer 41 of half separator 20A for air electrodes and half separator 20B for fuel gas electrodes which were set and done by the 3rd process C3 at the 4th process C4, The heat-resistant adhesives 22 of entering [ which has conductivity in this field ] a graphite are applied to homogeneity, and two half separators 20A and 20B in all are mutually stuck for these spreading sides. After that, the temperature up of these is carried out, they are heat-treated to 250-300 degrees C, after 2-hour heating, at 80 degrees C, with fixed planar pressure applied, and these two half separators 20A and 20B are joined.

[0061] The ribbed separator 2 to which two half separators 20A and 20B were joined is manufactured by low cost from the yield according to the above process [ 1st, 2nd, 3rd, and 4th ] C1, C2, C3, and C4.

[0062] Example 7. drawing 11 is drawing showing one example of the ribbed separator for fuel cells concerning invention of the 7th of this invention. The ribbed separator 2 of this example 7 is comparatively precise, sandwiches the thin carbon plate 23 (for example, amorphous new carbon film by NISSHINBO INDUSTRIES, INC.) with reinforcement by the mutual substantia-compacta carbon layer 41 side of half separator 20A for air electrodes of an example 1, and half separator 20B for fuel gas electrodes, through the heat-resistant adhesives 22, joins these and unifies. in addition, the thickness of this ribbed separator 2 -- the thickness of the half separators 20A and 20B -- respectively -- the thickness of 1.5mm and the carbon plate 23 -- 200 micrometers the thickness of the heat-resistant adhesives 22 by the side of both sides of the carbon plate 23 -- respectively -- 100 micrometers it is -- a sake -- 3.4mm -- becoming .

[0063] Although the ribbed separator 2 of this example 7 joins three members and it unifies, in order to insert the carbon plate 23 with the half separators 20A and 20B, flexural strength is strong and electric resistance and thermal resistance also become small compared with the ribbed separator 2 of the conventional comparable thickness on the property of the half separators 20A and 20B which consist of inclination carbon plates 30. Of course, reduction of laminating cost can also be aimed at.

[0064] In addition, although the amorphous new carbon film by NISSHINBO INDUSTRIES, INC. is used as a carbon plate 23 in this example 7, the thing of \*\* may be used as long as it is the substantia compacta not only in this but in high intensity.

[0065] Example 8. drawing 12 is drawing showing one example of the manufacture approach of the ribbed separator for fuel cells concerning invention of the 8th of this invention. This example 8 is related with the manufacture approach of the ribbed separator of the junction type of an example 7.

[0066] The 1st process D1 at which this manufacture approach manufactures the porous carbon plate of the fixed thickness which has the rib R for gas passageways in a whole surface side, The 2nd process D2 of said porous carbon plate which, on the other hand, forms the thin film of thermosetting resin in a side, The 3rd process D3 which heat-treats the porous carbon plate with which said thin film was formed, and changes said thin film into a substantia-compacta carbon layer with seal nature, The carbon plate 23 with thin reinforcement is inserted by the substantia-compacta carbon layer sides of the porous carbon plate of a pair with which said substantia-compacta carbon layer was formed, and it joins, and consists of the 4th process D4 which unifies these.

[0067] Since the 1st process D1 to the 3rd process D3 is the same as that of the 1st process A1 of an example 2 to 3rd process A3 respectively, the explanation is omitted. The field by the side of [ mutual ]

the substantia-compacta carbon layer 41 of half separator 20A for air electrodes and half separator 20B for fuel gas electrodes which were set and done by the 3rd process D3 at the 4th process D4, And after an organic solvent washes both sides of the carbon plate 23 which is thin and has reinforcement, the heat-resistant adhesives 22 are applied to homogeneity in these fields, and two half separators 20A and 20B in all and carbon plates 23 of each other are stuck for these spreading sides. Then, at 80 degrees C, a temperature up is carried out to 250-300 degrees C after 2-hour heating, it heat-treats, applying fixed planar pressure for these, and these two half separators 20A and 20B are joined.

[0068] The ribbed separator 2 to which the carbon plate 23 was joined between two half separators 20A and 20B is manufactured by low cost from the yield according to the above process [ 1st, 2nd, 3rd, and 4th ] D1, D2, D3, and D4.

[0069]

[Effect of the Invention] Since this invention is constituted as mentioned above, it does so effectiveness which is indicated below.

[0070] In the ribbed separator for fuel cells with which according to invention of the 1st of this invention a laminating is carried out to the both sides holding an electrolyte of a matrix by turns [ a cell and by turns ] with an electrode, and the rib for gas passageways is formed in those both sides The ingredient is really with the substantia-compacta carbon layer which is thin and has seal nature in the whole surface side of the porous carbon layer of fixed thickness constituted from two formal inclination carbon plates. Since [ of each porous carbon layer of two porous carbon plates ] it forms in a side on the other hand, where the engine performance and quality are fulfilled for the rib for those gas passageways, the electric resistance and thermal resistance of this ribbed separator can be stopped small, and reduction of laminating cost can be aimed at.

[0071] In the manufacture approach of the ribbed separator for fuel cells that according to invention of the 2nd of this invention a laminating is carried out to the both sides holding \*\* and an electrolyte of a matrix by turns [ a cell and by turns ] with an electrode, and the rib for gas passageways is formed in those both sides The 1st process which manufactures the porous carbon plate of the fixed thickness which has a rib for gas passageways in a whole surface side, Since it has the 2nd process of a porous carbon plate which, on the other hand, forms the thin film of thermosetting resin in a side, and the 3rd process which heat-treats the porous carbon plate with which the thin film was formed, and changes a thin film into a substantia-compacta carbon layer with seal nature, The ribbed separator concerning the 1st invention can be manufactured with the sufficient yield by low cost.

[0072] In the ribbed separator for fuel cells with which according to invention of the 3rd of this invention a laminating is carried out to the both sides holding an electrolyte of a matrix by turns [ a cell and by turns ] with an electrode, and the rib for gas passageways is formed in those both sides Since the ingredient is really with the substantia-compacta carbon layer which is thin and has seal nature between the porous carbon layers of the pair of fixed thickness constituted from a formal inclination carbon plate and the rib for the gas passageways is formed in both the external surface side of the porous carbon layer of an inclination carbon plate, In this ribbed separator, electric resistance and thermal resistance can be stopped still smaller from the ribbed separator concerning the 1st invention, and further reduction of laminating cost can be aimed at.

[0073] In the manufacture approach of the ribbed separator for fuel cells that according to invention of the 4th of this invention a laminating is carried out to the both sides holding an electrolyte of a matrix by turns [ a cell and by turns ] with an electrode, and the rib for gas passageways is formed in those both sides The 1st process which manufactures the porous carbon plate of the fixed thickness which has a rib for gas passageways in a whole surface side, While piling up and heat-treating the thin film sides of the porous carbon plate of a pair with which the thin film was formed and uniting these with the 2nd process of a porous carbon plate which, on the other hand, forms the thin film of thermosetting resin in a side Since it has the 3rd process which changes a thin film into a substantia-compacta carbon layer with seal nature, the yield is good and the ribbed separator concerning the 3rd invention can be manufactured by low cost.

[0074] In the ribbed separator for fuel cells with which according to invention of the 5th of this

invention a laminating is carried out to the both sides holding an electrolyte of a matrix by turns [ a cell and by turns ] with an electrode, and the rib for gas passageways is formed in those both sides The ingredient is really with the substantia-compacta carbon layer which is thin and has seal nature in the whole surface side of the porous carbon layer of fixed thickness constituted from two formal inclination carbon plates. Since the rib for the quantities of gas flow is formed in both the external surface side of the porous carbon layer of two inclination carbon plates which joined substantia-compacta carbon layer sides and were unified, In this ribbed separator, while being able to have the same effectiveness as the ribbed separator concerning the 1st invention, reduction of laminating cost can be aimed at from the ribbed separator concerning this 1st invention.

[0075] In the manufacture approach of the ribbed separator for fuel cells that according to invention of the 6th of this invention a laminating is carried out to the both sides holding an electrolyte of a matrix by turns [ a cell and by turns ] with an electrode, and the rib for gas passageways is formed in those both sides The 1st process which manufactures the porous carbon plate of the fixed thickness which has a rib for gas passageways in a whole surface side, The 2nd process of a porous carbon plate which, on the other hand, forms the thin film of thermosetting resin in a side, The 3rd process which heat-treats the porous carbon plate with which the thin film was formed, and changes a thin film into a substantia-compacta carbon layer with seal nature, Since it has the 4th process which joins the substantia-compacta carbon layer sides of two porous carbon plates with which the substantia-compacta carbon layer was formed, and is unified, the yield is good and the ribbed separator concerning the 5th invention can be manufactured by low cost.

[0076] In the ribbed separator for fuel cells with which according to invention of the 7th of this invention a laminating is carried out to the both sides holding an electrolyte of a matrix by turns [ a cell and by turns ] with an electrode, and the rib for gas passageways is formed in those both sides The ingredient really with the substantia-compacta carbon layer which is thin and has seal nature in the whole surface side of the porous carbon layer of fixed thickness Two formal inclination carbon plates, Although the carbon plate was inserted by the substantia-compacta carbon layer sides of two inclination carbon plates and coalesce junction was carried out, since it constitutes from a carbon plate with thin reinforcement, and the rib for quantities of gas flow is formed in porous carbon \*\*\*\*\* external surface, The reinforcement of the simple substance of a ribbed separator can be raised, and reduction of laminating cost can be aimed at.

[0077] In the manufacture approach of the ribbed separator for fuel cells that according to invention of the 8th of this invention a laminating is carried out to the both sides holding an electrolyte of a matrix by turns [ a cell and by turns ] with an electrode, and the rib for gas passageways is formed in those both sides The 1st process which manufactures the porous carbon plate of the fixed thickness which has a rib for gas passageways in a whole surface side, The 2nd process of a porous carbon plate which, on the other hand, forms the thin film of thermosetting resin in a side, The 3rd process which heat-treats the porous carbon plate with which the thin film was formed, and changes a thin film into a substantia-compacta carbon layer with seal nature, A carbon plate with thin reinforcement is inserted by the substantia-compacta carbon layer sides of two porous carbon plates with which the substantia-compacta carbon layer was formed, and it joins, and since it has the 4th process which unifies these, the yield is good and the ribbed separator concerning the 7th invention can be manufactured by low cost.

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[Translation done.]